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Optical properties of tungsten thin films perforated with a bidimensional array of subwavelength holes

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We present a theoretical investigation of the optical transmission of a dielectric grating carved in a tungsten layer. For appropriate wavelengths tungsten shows indeed a dielectric behaviour. Our numerical simulations leads to theoretical results similar to those found with metallic systems studied in earlier works. The interpretation of our results rests on the idea that the transmission is correlated with the resonant response of eigenmodes coupled to evanescent diffraction orders.

For a few years, properties and technological applications of one- or two-dimensionnal metallic gratings have received a growing interest. In 1998, Ebbesen *et al* [1] reported on optical transmission experiments performed on periodic arrays of subwavelength cylindrical holes drilled in a thin metallic layer deposited on glass. These experiments renewed the motivation for investigating metallic gratings. Two attractive characteristics of their results are often cited : the transmission, which is a lot higher than the addition of individual holes contributions, and the peculiar wavelength dependance of the transmission. Further work [1-7] has suggested that these features arise from the presence of the metallic layer, and call for the presence of surface plasmons in order to explain these transmission characteristics. In particular, they have identified the convex high transmission regions, i.e., the regions between the minima, as regions dominated by the plasmon response. However, many questions remain and need to be answered in order to clarify the mechanisms involved in these experiments.

In a recent paper [8], extensive simulations have been performed in order to understand the optical properties of a chromium layer similar to those developed in experiments. Recalling of Wood's anomalies [9], it is shown that the transmission and reflection are better described as Fano's profiles correlated with resonant response of the eigenmodes coupled to nonhomogeneous diffraction orders. Indeed, as explained by V.U. Fano [10], A. Hessel and A.A. Oliner [11] for one dimensional gratings, Wood's anomalies are related to eigenmodes grating excitation. To be accurate, they have shown that Wood's anomalies [11] may arise in two ways. The first case occurs at Rayleigh's wavelengths, when a diffracted order becomes grazing to the grating plane [12]. The diffracted beam intensity then increases just before the diffracted order vanishes. The other case is related to a resonance effect. Such resonances come from coupling between the incident light and the eigenmodes of the grating. Both types of anomalies may occur separately and independently, or may almost coincide.

In our previous paper [8], as we used metal in our device, it seemed natural to assume that these resonances are surface plasmons. Nevertheless, it is important to note that our analysis did not make any hypothesis on the origin of the eigenmodes. This implies that it could be possible to obtain transmission curves similar to those found for metals, by substituting guided modes or other types of polaritons to the surface plasmons. This would allow, for instance, to substitute dielectric guided modes to metallic excitations. This is the purpose of the present paper. We could also use films made of ionic crystal in the far infrared, and deal with phonon-polaritons (in progress).

In this paper we perform simulations of a device which consists of arrays of subwavelength cylindrical holes in a tungsten layer deposited on glass substrate (figure 1). Indeed, tungsten becomes dielectric on a restricted domain of wavelength in the range $240 - 920 \text{ nm}$, i.e. the real part of its permittivity becomes positive. The permittivity values are taken from experiments [13]. So, whereas plasmons cannot exist, we show that the transmission pattern is similar to that obtained in the case of a metallic film. However, experiment has also shown that a germanium film is not able to give rise to an Ebbesen effect. We then also need to explain here why in the case of germanium nothing interesting can be observed.

Our simulations rest on a coupled modes method (which takes into account the periodicity of the device permittivity) associated with the use of the scattering matrix formalism [8,14]. In this way, we calculate the amplitudes of the reflected and transmitted field, for each diffracted order (which correspond to a vector \vec{g} of the reciprocal lattice) according to their polarization (s or p). In the following, for a square grating of parameter a , note that, $\vec{g} = \frac{2\pi}{a}(i, j)$, so that the pair of integers (i, j) denotes the corresponding vector of the reciprocal lattice (i.e. diffraction order). In addition, we recall that Rayleigh's wavelengths are defined as

$$\lambda_R^{u,i,j} = a\sqrt{\varepsilon_u}(i^2 + j^2)^{-\frac{1}{2}} \quad (1)$$

where ε_u represents either the permittivity of the vacuum (ε_v), or of the dielectric substrate (ε_d). We calculate the zero transmission order, referring to the experimental measurements performed in the metallic case. We can also estimate the partial density of states (PDOS) for some positions in the first Brillouin's zone.

The calculated transmission of the incident wave is shown against wavelength in Fig. 2a for the zero diffrac-

tion order, and for incident light normal to the surface of a W film on glass. The holes diameter is set to $d = 320$ nm and the thickness of the film is $h = 100$ nm. The transmission is given for square gratings of parameter $a = 550$ nm, 500 nm and 450 nm, respectively. In Fig. 2a, it is shown that the transmission increases with the wavelength, and that it is characterized by minima marked 1 and 2 on the figure, which are located closer after Rayleigh's wavelength $\lambda_R^{d,0,\pm 1}$ and $\lambda_R^{v,0,\pm 1}$, respectively. The locations of these minima are shifted toward larger wavelengths when the grating size increases. Our investigations show that these minima disappear when considering a system without hole. Note that these results are qualitatively similar to those from the experimental data of Ebbesen *et al* in the case of metallic layers [1,2].

Fig 2b shows the calculated zero-order transmission as a function of the incident wavelength, for gratings with different holes diameters. As in the case of metallic gratings [1,2,6,7], it is clear that transmission curves does not depend of the hole diameter except for the amplitude of the spectral features.

In order to underline the correlation between the transmission amplitude and the holes diameter we also give in Fig. 2c the transmission amplitude for many hole diameter values (dots in Fig. 2c). The transmission values are those which correspond to the maximum located at 576.7 nm for a square grating of parameter $a = 500$ nm, with the chosen thickness of the film ($h = 100$ nm). We show that the transmission amplitude increases exponentially as a function of the hole diameter (solid line in Fig. 2c). This result suggests that there is no cavity mode involved in those phenomena.

In Fig 2d we give the calculated transmission as a function of the wavelength of the incident wave for the zeroth diffraction order, for different layer thicknesses. As in the case of metallic gratings [1,2,6,7], we show that the behaviour of the transmission curves does not depend of the thickness except for the overall amplitude.

The results we shown in Fig 2 present behaviours very similar to those obtained with metals cases [1,2], even though we use tungsten in its dielectric domain. Let us discuss this point. As explained in [8], the eigenmodes of the grating play a crucial role in those experiments. Let us first recall that, reflected and transmitted amplitudes are linked to the incident field through the S scattering matrix. Let us define F_{scat} as the scattered field, and F_{in} as the incident field. Then, F_{scat} is related to F_{in} via the scattering matrix, in such a way that

$$S(\lambda)F_{in}(\lambda) = F_{scat}(\lambda) \quad (2)$$

In this way, the eigenmodes of the structure are solution of eq.2 which exist in absence of source, i.e. when

$$S^{-1}(\lambda_\eta)F_{scat}(\lambda_\eta) = 0 \quad (3)$$

This homogeneous problem is well known in the theory of gratings [8,11]. Complex wavelengths $\lambda_\eta = \lambda_\eta^R + i\lambda_\eta^I$, for which eq.3 has non-trivial solutions require that

$$\det(S^{-1}(\lambda_\eta)) = 0 \quad (4)$$

so that they coincide with the poles of the complex function $\det(S(\lambda))$.

If we extract the singular part of S corresponding to the eigenmodes of the structure, we can write S as [8,11]

$$S(\lambda) = \sum_{\eta} \frac{R_{\eta}}{\lambda - \lambda_{\eta}} + S_h(\lambda) \quad (5)$$

i.e. under the form of a generalized Laurent series, where R_{η} is the residue associated with the pole λ_{η} and $S_h(\lambda)$ is the holomorphic part of S which corresponds to physically non-resonant process.

Thus, assuming that $f(\lambda)$ is the m^{th} component of $F_{scat}(\lambda)$, we have, for the expression of $f(\lambda)$ in the neighborhood of one pole λ_{η} [8,10,11]

$$f(\lambda) = \frac{r_{\eta}}{\lambda - \lambda_{\eta}} + s(\lambda) \quad (6)$$

where $r_{\eta} = [R_{\eta}F_{in}]_m$ and $s(\lambda) = [S_h(\lambda)F_{in}]_m$.

We consider the case where non-resonant processes cannot be neglected, and assume that $s(\lambda) \sim s_0$ is a constant value. Thus, it is easy to show that eq.6 can be written as [8,11]

$$|f(\lambda)|^2 = \frac{(\lambda - \lambda_z^R)^2 + \lambda_z^I^2}{(\lambda - \lambda_{\eta}^R)^2 + \lambda_{\eta}^I^2} |s_0|^2 \quad (7)$$

with

$$\lambda_z^R = \lambda_{\eta}^R - \nu^R \quad \text{and} \quad \lambda_z^I = \lambda_{\eta}^I - \nu^I \quad (8)$$

where

$$\nu = \frac{r_{\eta}}{s_0} \quad (9)$$

Coefficient ν measures the significance of the resonant effect, compared with non-resonant contributions. $\lambda_z = \lambda_z^R + i\lambda_z^I$ denotes the zero of eq.6 and 7. This last expression take into account the interferences between resonant and non-resonant processes, which lead to asymmetric transmission profiles. Eq.7 leads to a typically resonant process (described by a lorentzian curve) or to a typical asymmetric behavior where a minimum is following a maximum, and *vice versa* assuming the values of ν . We note that this properties, which results from the interference between resonant and non-resonant processes, are similar to those described by A.A. Hessel, A. Oliner [11] and V.U. Fano [10]. The profiles like those describe on figure 3a are often referred to as "Fano's profiles".

It is now necessary to find the structure eigenmodes. To this purpose, we show in Fig. 3b the partial density of states, i.e. the density of states in the center Γ of the first Brillouin's zone. Indeed, when the incident light is normal to the surface, our assumption is that the DOS at Γ is the dominant contribution to those phenomena.

The solid line shows the partial DOS which is calculated in the case of a grating with $d = 320 \text{ nm}$, $h = 100 \text{ nm}$ and $a = 500 \text{ nm}$. The dashed line shows the partial DOS which is calculated for a similar system in absence of holes. The overall pattern of the partial DOS of the grating is related to the pattern of the partial DOS of the planar tungsten layer. It appears some sharp localized peaks related to the eigenmodes of the tungsten grating. Such eigenmodes can only be associated with guided modes of the tungsten grating assuming its dielectric properties. We note the Rayleigh's anomalies (circles around sharp minima of DOS) which appear just on the side of the eigenmodes at shorter wavelength. We also show the corresponding transmission for sake of comparison. One clearly notices the absence of coincidence between the position of the eigenmodes and the transmission maxima. Nevertheless the eigenmodes are behind of the typical profiles of the transmission curves [8]. In fact, one can interpret the features lineshapes as arising from resonant Wood's anomalies, similar to those studied by V. U. Fano [10] and by A. Hessel and A.A. Oliner [11].

Let us consider an incident propagative wave, which diffracts and generates a evanescent diffraction order. Such an order is coupled with a guided eigenmode which is characterized by a complex wavelength λ_η . It becomes possible to excite this eigenmode which lead to a feedback reaction on the evanescent order. For this reason, A. Hessel and A.A. Oliner called such order a "resonant order". This process is related to the resonant term.

The evanescent resonant order can diffract due to the layer corrugation and generates a contribution to the propagative zero diffraction order. Thus, one can ideally expect to observe a resonant (lorentzian) profile for the zero diffraction order. Nevertheless, it is necessary to take into account a nonresonant diffraction process related to the holomorphic term of the scattering matrix. So, the incident wave, generates a propagative zero order. Then, one takes into account the interference of two rates, i.e. resonant and non resonant contribution to order zero. It appears profiles which are typically the Fano's profiles which correspond to resonant process where one takes into account nonresonant effects. One note that a maximum in transmission does not necessary correspond to the maximum of resonance of a diffraction order. So, the Fano's profiles behaviour of the transmission, result from the superimposing of resonant and nonresonant contributions to the zero diffraction order.

If one refers to the present situation the resonance is close to Rayleigh's wavelength. Consequently, asymmetric Fano's profiles are shifted toward Rayleigh's wavelength in the same way. Then the transmission minima, which in fact correspond to minima of Fano's profiles, disappears when crossing a Rayleigh's wavelength. Maxima of the transmission, which is shifted towards larger wavelengths as shown in [8], correspond to the maxima of Fano's profiles.

Then, the maxima do not correspond explicitly to maxima of resonances but rather to the maxima of Fano's

profiles originating from the excitation of guided-modes and not surface plasmons as in metallic cases [8]. Nevertheless, it is necessary to emphasize why in the case of germanium, which is a dielectric, nothing similar has been observed. In fact, in the wavelength domain of our simulations the imaginary part of the tungsten permittivity is large, whereas in the case of germanium, in the domain wavelength studied by Ebbesen *et al*, the imaginary part of the permittivity is weak enough to consider germanium as transparent. In such situation, the contribution of the holomorphic part of the scattering matrix is much larger than the resonant part so that the latter can be neglected. Thus one completely loses the benefit of the resonant effects which plays a fundamental role in the transmission lineshapes. Moreover, in the case of tungsten, the opacity is such that there is not enough direct transmitted light to mask the effects of resonant processes.

It would be desirable to obtain more experimental data about tungsten gratings, as a way to confirm our theoretical findings. The ability to use dielectric layers would open a potential of Ebbesen's devices. One notes that the role of guided resonances and Fano's profiles in photonic crystal slabs have been also underlined by S. Fan and J. D. Joannopoulos in a recent paper [15].

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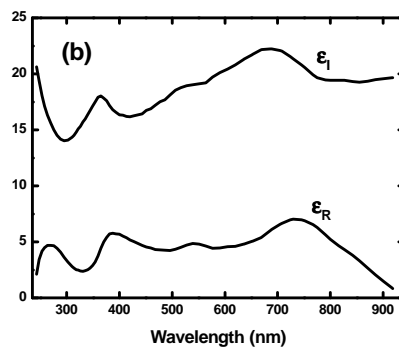
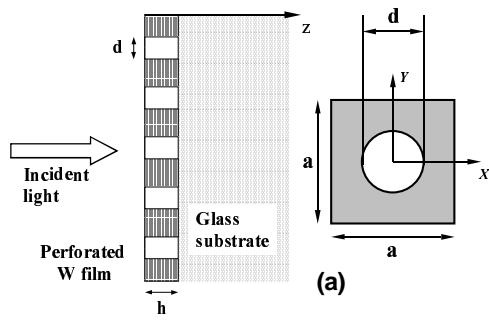
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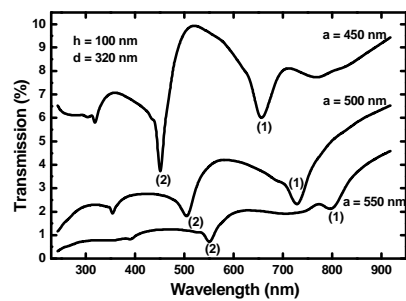
CAPTIONS

FIG. 1. (a) Diagrammatic view of the system under study. Transmission is calculated for the zeroth order and at normal incidence as in experiments. (b) Real and imaginary part of tungsten permittivity.

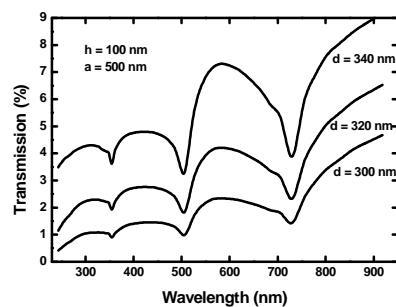
FIG. 2. Percentage transmission of the incident wave on the surface against wavelength, for the zeroth diffraction order, for different parameters of the square grating (a), for different hole diameters (b) and for different thickness (d). Part (c) represent the transmission against hole diameter for a given wavelength.

FIG. 3. (a) Representation of typical Fano's profiles. (b) PDOS against wavelength compare with transmission.

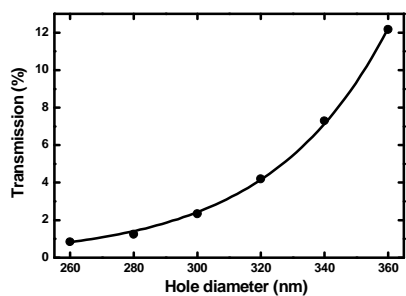




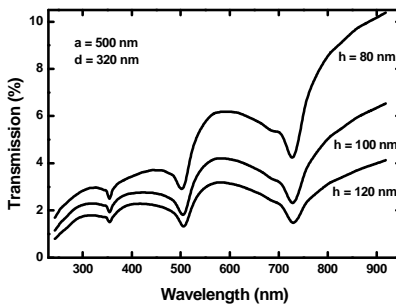
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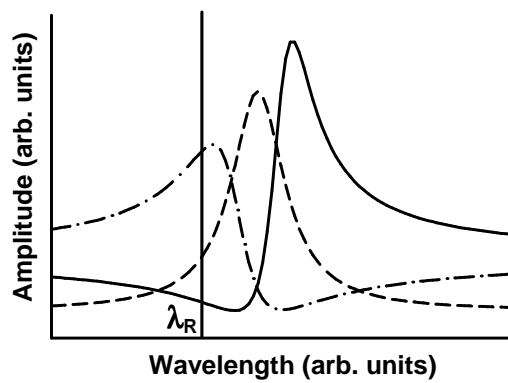
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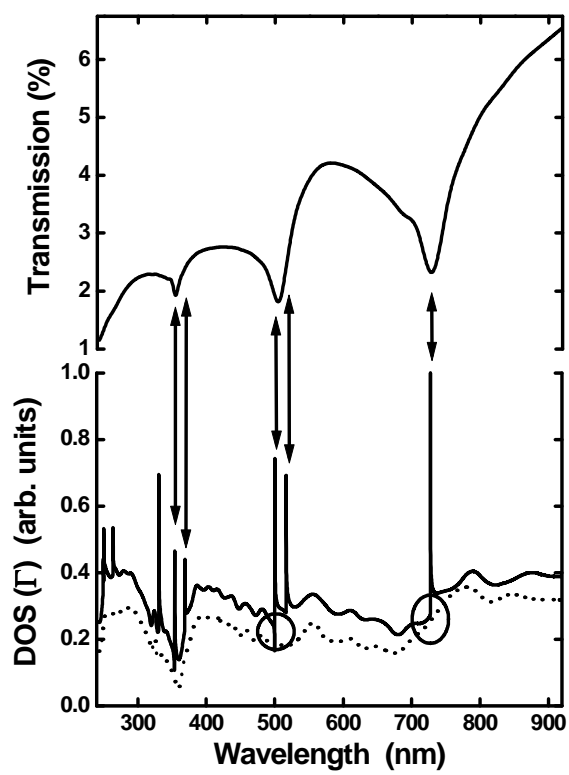
(c)



(d)



(a)



(b)